

BERYLLIUM-7 AND LEAD-210 IN THE ATMOSPHERE AND SURFACE SNOW OVER THE GREENLAND ICE SHEET IN THE SUMMER OF 1989

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**Abstract.** The concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  were measured in surface air and fresh and aging snow samples from Summit ( $72^\circ 20'\text{N}$ ,  $38^\circ 45'\text{W}$ ) and Dye 3 ( $65^\circ 10'\text{N}$ ,  $44^\circ 45'\text{W}$ ) Greenland, during June and July 1989. The aerosol concentrations of these radionuclides showed rapid variations at both sites, but were nearly twice as high, on average, at Summit. Concentrations in the 16 fresh snowfall events that were sampled also showed wide variability, but the averages were the same at the two sites. The apparent difference in air-snow fractionation and the lack of coherence in the concentration in air time series between the two sites indicate previously unsuspected complexity in atmospheric dynamics over the ice sheet. Improved understanding of atmospheric processes, and how the results of those processes are recorded in snow and ice, is crucial for full interpretation of the information about past atmospheric chemistry and climate contained in the snow and ice of glaciers around the world.

Introduction

In this era of concern over anthropogenically induced global climate change there is an urgent need to improve our understanding of the atmosphere-ocean-climate system. Records preserved in the snow and ice of the polar ice sheets provide high-resolution temporal information on past climatic and atmospheric conditions.

It is well established that polar ice records large-scale changes in atmospheric chemistry like those resulting from steadily increasing anthropogenic emissions [Boutron and Delmas, 1980; Neftel et al., 1985; Mayewski et al., 1986; Peel, 1986; Boutron and Patterson, 1986; Finkel et al., 1986; Steffensen, 1988] and circulation patterns [Dansgaard et al., 1989; Petit et al., 1990], but the details of the relationship between the chemistry of present day snow and the atmosphere are unclear. The similar, if not greater, lack of understanding of modifications of snow chemistry by early postdepositional and firmification processes limit the confidence with which glaciochemical records can be interpreted as direct proxy records of atmospheric chemistry.

Although numerous detailed snow chemistry and atmospheric sampling campaigns have been conducted in the polar regions over the past 30 years or so, the first extended program focusing on air-snow relationships and postdepositional changes in snow chemistry did not begin until 1988. The yearlong (August 1988 through July 1989) Dye 3 gas, aerosol, and snow sampling program (DGASP) involved the collection of nearly continuous aerosol samples, weekly gas samples, and numerous fresh and aged snow samples on the Greenland Ice Sheet. Provisions were also made to allow resampling of temporally well defined intervals of the snowpack over the next several years. The atmospheric sampling component of the Greenland Ice Sheet Project Two

(GISP 2), which began in the 1989 field season, has objectives similar to DGASP but differs from it in two important aspects. The lack of a permanent manned station in the Summit region of Greenland limits sampling to the mid-May to mid-September summer season. On the other hand, the remoteness of this site greatly reduces potential problems of local pollution, which imposed limitations on sample collection at Dye 3. For the GISP 2 atmospheric sampling program every effort has been made to take full advantage of the relatively pristine Summit region. Atmospheric and snow sampling is conducted at a small, solar powered camp (ATM) located about 30 km into the "clean air" sector that was established around the drill camps of GISP 2 and the companion European effort (Greenland Ice Core Project (GRIP)) (bearing from GISP 2 to ATM  $204^\circ$  true) (Figure 1).

The broad scientific objective of the atmospheric sampling at Dye 3 and Summit is to examine the sources, transport mechanisms, depositional processes, and early postdepositional changes influencing the incorporation and preservation of atmospheric constituents in the snow and firm of the Greenland Ice Sheet. The basic approach was to make measurements of a suite of atmospheric constituents (both gas phase and those associated with aerosols) at the highest feasible temporal resolution, determine the concentrations of the same species in freshly fallen snow, and then monitor the snow chemistry over time (days to years). Ancillary meteorological and climatological observations should help elucidate the causes of temporal changes in the relative importance of the various sources, transport pathways, atmospheric removal mechanisms, and diagenetic processes impacting snowpack chemistry.

Measurements of the atmospheric radionuclides  $^7\text{Be}$  and  $^{210}\text{Pb}$  were included in both DGASP and the first year of the GISP 2 atmospheric sampling program. In the atmosphere these radionuclides are predominantly associated with submicron aerosols [Maenhaut et al., 1979; Bondietti et al., 1987, 1988], so they provide general information on the depositional and early postdepositional processes impacting these aerosols and their associated chemical constituents. The altitudinally distinct sources of cosmogenic  $^7\text{Be}$  (cosmic ray induced spallation of N and O in the atmosphere, with maximum production at about 15 km [Bhandari et al., 1970]) and the  $^{238}\text{U}$  series  $^{210}\text{Pb}$  (almost strictly continental, predominantly from the emanation of gaseous  $^{222}\text{Rn}$ , [Turekian et al., 1977] suggest that they may allow assessment of the relative importance of stratospheric and tropospheric transport pathways in bringing continentally derived material to the Greenland Ice Sheet.

This paper focuses on the results of the first sampling season at the GISP 2 ATM, with comparisons to DGASP for the two months when both projects operated simultaneously. Analysis and interpretation of the complete DGASP data set are currently in progress and will be presented in forthcoming papers.

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Methods

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Aerosol samples were collected on Whatman GF/A glass fiber filters coupled to high-volume vacuum pumps with 2"

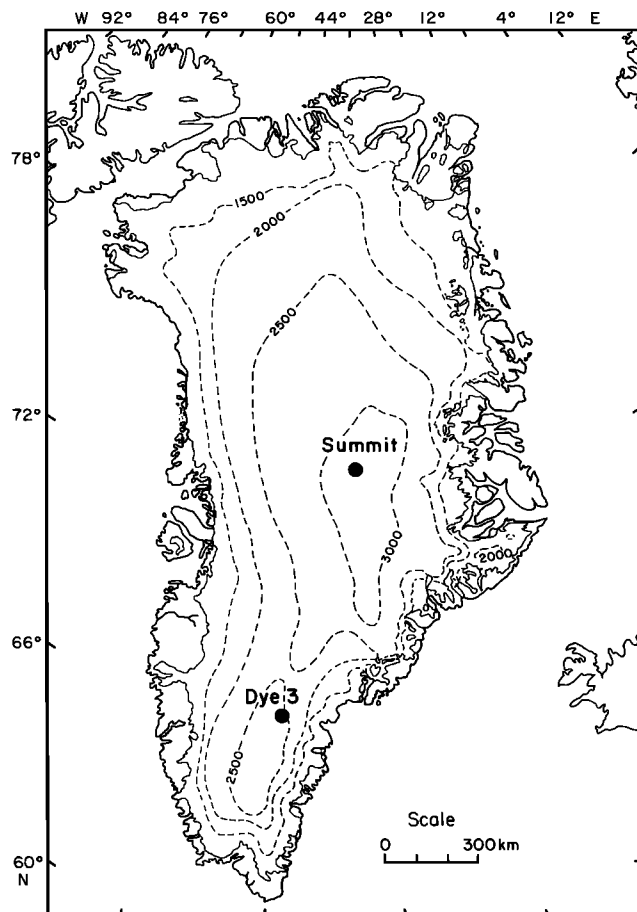


Fig. 1. Map of Greenland showing the sampling sites at Summit and Dye 3. Contours show surface elevation in meters above sea level.

PVC sampling lines. Sampled air volumes were determined by in-line dry gas meters. Corrections for temperature and the reduced pressure in the sampling line allowed conversion of the measured volumes to standard cubic meters ( $\text{m}^3$  STP). At ATM,  $20 \times 25 \text{ cm}^2$  filters were used, giving a face velocity of about  $0.2 \text{ m s}^{-1}$  at a flow rate of  $30\text{--}35 \text{ m}^3 \text{ STP hour}^{-1}$ . Four 47-mm-diameter filters were exposed concurrently at Dye 3, yielding a face velocity of about  $0.5 \text{ m s}^{-1}$  at a flow rate of approximately  $13 \text{ m}^3 \text{ STP hour}^{-1}$ .

Between June 3 and July 24, 1989, 37 aerosol samples were collected at Summit. A total of 62 potential sampling hours, in 6 blocks of 12 hours or less, were lost due to insufficient generation of electrical power during extended cloudy periods. At Dye 3, 17 aerosol samples were collected during roughly the same period (June 2 to July 28).

Fresh snow was collected after every event which produced observable accumulation (11 at Summit, 5 at Dye 3). Snow was sampled by carefully scraping the newly deposited surface layer into polyethylene bags with clean Plexiglas scrapers. At ATM the area sampled, new layer depth, and sample mass were recorded for all except one sample; usually, only the sample mass was noted at Dye 3. Two traverses were conducted to assess spatial variability of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in fresh snow in the Summit region. Snow samples for radionuclide determinations were melted just enough to ensure that the bags were not going to leak, then 0.5 mL of concentrated HCl and a stable Be tracer were added. The spiked samples remained in the bags at least overnight before being transferred to precleaned 1-L polyethylene bottles for transport to the University of New Hampshire (UNH).

Upon receipt at the UNH laboratory, aerosol filters were pressed into 4-mL polyethylene vials for analysis. Snow samples were spiked with a stable Cl tracer, then concentrated by sub-boiling evaporation in Teflon beakers until they could be quantitatively transferred to the 4-mL vials. (The Be and Cl tracers were added to check the yield of subsequent chemical treatments required to prepare the samples for accelerator mass spectrometric determinations of  $^{10}\text{Be}$  and  $^{36}\text{Cl}$ . The snow and aerosol samples discussed here are being analyzed for these additional cosmogenic radionuclides by R. Finkel of Lawrence Livermore National Laboratory.)

$^7\text{Be}$  and  $^{210}\text{Pb}$  concentrations were determined by direct gamma spectrometry, using a germanium well detector connected to a microprocessor-based multichannel analyzer. Calibration of the counting system was accomplished by preparing NBS traceable standard solutions in geometries identical to the samples (two different filter standards and one liquid standard). These standards were recounted at approximately monthly intervals to verify the stability of the counting system.

## Results

### Aerosols

The time series of  $^7\text{Be}$  and  $^{210}\text{Pb}$  concentrations in ground level air at Summit and Dye 3, Greenland, exhibited substantial short-term variability (Figure 2). The Summit data demonstrate this more clearly due to the higher sampling frequency.  $^7\text{Be}$  and  $^{210}\text{Pb}$  concentrations were consistently higher at Summit than Dye 3. The ratios, Summit/Dye 3, of volume-weighted average concentration of  $^7\text{Be}$  were 1.9 and 1.6 in June and July, respectively, while the corresponding ratios for  $^{210}\text{Pb}$  were 1.8 and 1.9. It appears that this is a real difference in aerosol concentrations, since the glass fiber filter media has been shown to capture 0.1- to 0.3-micron aerosols with >99.9% efficiency over the face velocity range  $0.15\text{--}1.33 \text{ m s}^{-1}$  [Liu et al., 1984]. Surprisingly, there was high correlation between  $^7\text{Be}$  and  $^{210}\text{Pb}$  concentrations at both Summit ( $r=0.55$ , significant at  $p=0.001$ ) and Dye 3 ( $r=0.68$ , significant at  $p=0.005$ ). However, there does not appear to be much coherence in the short-term trends of the time series between the two sites (Figure 3).

### Snow

The concentrations of the atmospheric radionuclides in fresh snow at the two sites during June and July varied nearly thirtyfold for  $^7\text{Be}$  and sixtyfold for  $^{210}\text{Pb}$  (Table 1). However, the average concentrations in snow were quite similar, despite the nearly twofold higher concentrations in the atmosphere over Summit. The snow samples collected in the last 2 months of DGASP averaged  $18.7 \pm 7.9 \text{ pCi } ^7\text{Be kg}^{-1}$  and  $1.59 \pm 1.24 \text{ pCi } ^{210}\text{Pb kg}^{-1}$ , while the first seven GISP 2 samples (collected through July 10) averaged  $16.8 \pm 11.1 \text{ pCi } ^7\text{Be kg}^{-1}$  and  $1.08 \pm 0.97 \text{ pCi } ^{210}\text{Pb kg}^{-1}$ .

The GISP 2 data presented in Table 1 include only the snow samples collected at ATM from events where the newly deposited snow was present as an identifiable, relatively undisturbed, layer. In addition, two fresh snow sampling traverses were conducted to examine spatial variability of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in the Summit region (Table 2). The snow sampled on May 30 fell during the preceding 24 hours, accompanied by high wind and blowing snow conditions, while on June 24 the snow accumulated in nearly still air conditions. Comparison of these data indicates that the redistribution of newly fallen snow by wind, and its mixing with variable amounts of older snow, greatly increases the heterogeneity of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in the surface snow.

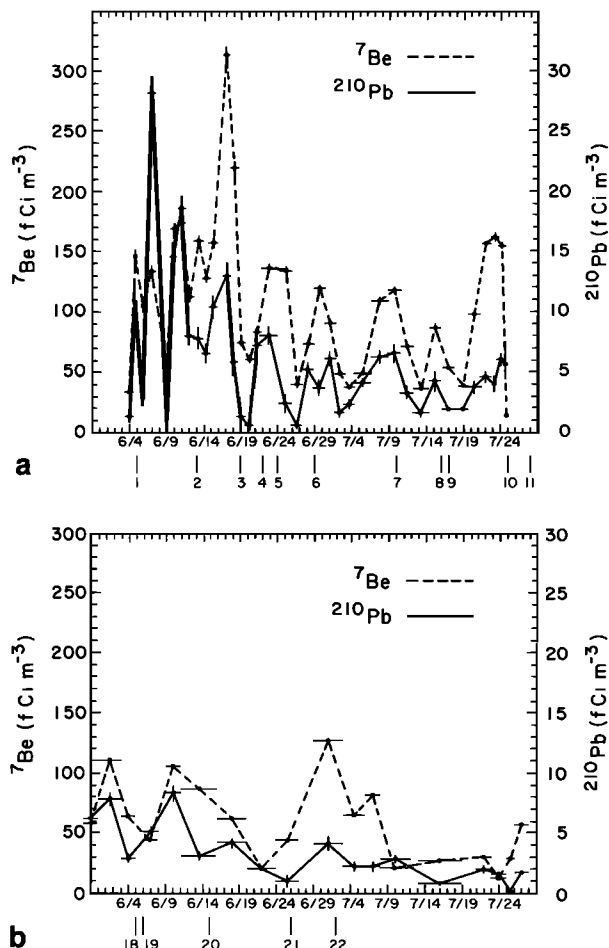


Fig. 2. Concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in surface level air over the Greenland Ice Sheet at (a) Summit and (b) Dye 3 during the summer of 1989. The numbered vertical lines beneath each time series indicate snowfall events that were sampled during the study period.

Throughout the first GISP 2 field season rime ice, or frost deposits were collected on vertical screens at the GISP 2 and ATM camps for stable isotope studies (P. Grootes and J. White, principal investigators). On July 4 and 5 the mass deposited on the ATM screen was large enough so that material was also available for  $^7\text{Be}$  and  $^{210}\text{Pb}$  assay. On July 4 the rime or frost contained  $30.4 \pm 1.8$  and  $4.86 \pm 0.55$  pCi kg<sup>-1</sup> of  $^7\text{Be}$  and  $^{210}\text{Pb}$ , respectively, and the July 5 concentrations were  $32.6 \pm 2.7$  and  $4.83 \pm 0.74$  pCi kg<sup>-1</sup>. The  $^7\text{Be}$  levels are slightly higher than those observed in fresh snowfall on the nearest sampling dates, while the  $^{210}\text{Pb}$  levels are notably higher (Table 1), in accord with previous studies showing that aerosol-associated major ions are concentrated in rime relative to snow [Borys et al., 1983; Mitchell and Lamb, 1989].

Discussion

Aerosol Concentrations

The aerosol concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  presented above appear to be the first for stations on the ice in central Greenland. The average  $^7\text{Be}$  concentrations at Summit during June and July 1989 were considerably (1.4-4.9 times) higher than those that have been observed at coastal stations in Greenland and Barrow, Alaska, while at Dye 3 the  $^7\text{Be}$  concentrations were higher than those seen at all of the Arctic locations except Thule (Table 3). In contrast, the  $^7\text{Be}$  concentrations over the Greenland Ice Sheet were lower than those that have been reported for the summer months at several Antarctic stations (Table 3).  $^{210}\text{Pb}$  concentrations in the aerosol over Greenland were comparable to those reported in previous work at Barrow and slightly higher than the usual summer values in Antarctica (Table 3).

The higher  $^7\text{Be}$  concentrations in the aerosols over the Greenland Ice Sheet, compared to low elevation, coastal, Arctic sites, presumably reflects the higher elevations of the ice sheet stations. However, Viezee and Singh [1980] reported little or no variation of  $^7\text{Be}$  concentrations over the altitude range 23-4767 m at four continental sites (near 9°N, 75°W), which suggests that the presence of the Greenland Ice Sheet as a very abrupt, high obstacle may facilitate the mixing of  $^7\text{Be}$ -rich air from the upper troposphere and/or stratosphere

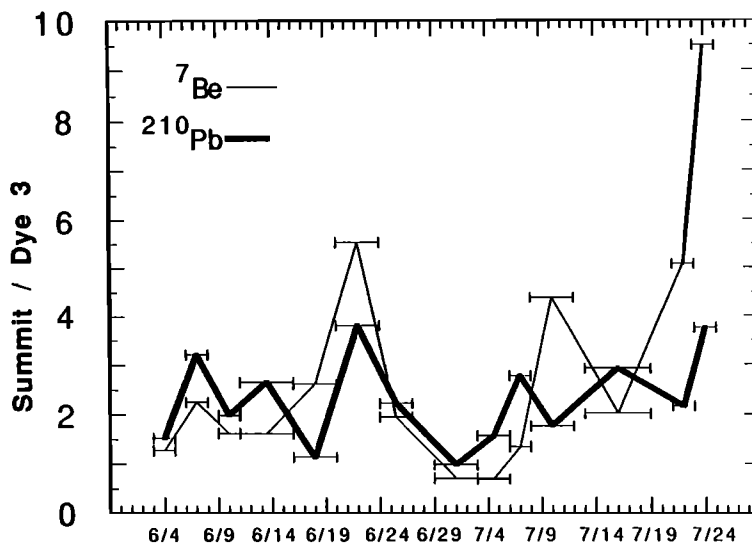


Fig. 3. Values of the ratios (Summit/Dye 3) of the concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  plotted against time. The concentrations at Summit were averaged to match the longer sampling intervals at Dye 3.

TABLE 1. Atmospheric Radionuclides Deposited in Fresh Snow at Two Sites in Greenland

| Date<br>1989                     | Concentration, pCi/kg |                   | Scavenging Ratio <sup>a</sup> |                   | Deposition, pCi/m <sup>2</sup> |                   |
|----------------------------------|-----------------------|-------------------|-------------------------------|-------------------|--------------------------------|-------------------|
|                                  | <sup>7</sup> Be       | <sup>210</sup> Pb | <sup>7</sup> Be               | <sup>210</sup> Pb | <sup>7</sup> Be                | <sup>210</sup> Pb |
| <u>Summit (72°20'N, 38°45'W)</u> |                       |                   |                               |                   |                                |                   |
| June 5                           | 3.44±0.61             | nd                | 35                            | ---               | 13.67±2.42                     | nd                |
| June 13                          | 6.31±0.60             | 1.36±0.20         | 51                            | 217               | 14.59±1.38                     | 3.15±0.47         |
| June 19                          | 13.85±0.78            | 1.34±0.19         | 195                           | 1083              | 49.44±2.78                     | 4.79±0.69         |
| June 22                          | 24.91±1.78            | 1.49±0.44         | 191                           | 195               | 13.28±0.95                     | 0.80±0.23         |
| June 24                          | 34.45±1.46            | 2.17±0.29         | 267                           | 940               | 40.74±1.73                     | 2.56±0.34         |
| June 29                          | 11.67±0.75            | 0.51±0.11         | 103                           | 143               | 54.93±3.54                     | 2.39±0.54         |
| July 10                          | 22.99±0.61            | 0.46±0.10         | 205                           | 73                | No Area Recorded               |                   |
| July 16                          | 2.31±0.38             | 1.45±0.36         | 45                            | 792               | 13.09±2.15                     | 8.22±2.04         |
| July 17                          | 1.20±0.30             | 0.05±0.06         | 23                            | 25                | 8.86±2.22                      | 0.37±0.44         |
| July 25                          | 4.62±0.67             | nd                | 366                           | ---               | 38.71±5.61                     | nd                |
| July 28                          | 6.91±0.75             | 3.01±0.38         | ns <sup>b</sup>               | ns <sup>b</sup>   | 22.47±2.44                     | 9.79±1.24         |
| <u>Dye 3 (65°10'N, 44°45'W)</u>  |                       |                   |                               |                   |                                |                   |
| June 5                           | 26.30±1.40            | 2.76±0.35         | 413                           | 968               | 115-175 <sup>c</sup>           | 12-18             |
| June 6                           | 14.66±1.48            | nd                | 329                           | ---               | 40-90                          | nd                |
| June 15                          | 13.68±0.70            | 1.10±0.18         | 159                           | 356               | 60-135                         | 5-11              |
| June 26                          | 10.83±0.82            | 2.95±0.34         | 245                           | 2875              | 15-45                          | 4-12              |
| July 2                           | 28.03±1.06            | 1.13±0.25         | 222                           | 276               | 145-305                        | 6-12              |

Here, nd means not detected.

<sup>a</sup>Scavenging ratios calculated as  $C_{sp}/C_a$ , where  $C_s$  is concentration in snow,  $C_a$  is aerosol concentration and  $\rho_a$  is the density of the air (Davidson et al., 1987).

<sup>b</sup>No aerosol samples collected for this time period.

<sup>c</sup>Estimates for Dye 3 very uncertain, see text for discussion.

TABLE 2. Atmospheric Radionuclides in Traverses of Fresh Snowfall Sampled Near Summit During Summer 1989

| Distance*, km                                   | Concentration, pCi/kg |                   | Deposition, pCi/m <sup>2</sup> |                   | Depth, cm |
|---|-----------------------|-------------------|--------------------------------|-------------------|-----------|
|   | <sup>7</sup> Be       | <sup>210</sup> Pb | <sup>7</sup> Be                | <sup>210</sup> Pb |           |
| <u>May 30, GISP 2 to GRIP, Bearing 90° True</u> |                       |                   |                                |                   |           |
| 2   | 0.56±0.42             | 1.62±0.21         | 12.07±9.05                     | 34.90±4.53        | 12.5      |
| 7   | 4.42±0.45             | 2.49±0.22         | 80.61±8.21                     | 45.41±4.01        | 10.0      |
| 12  | 9.32±0.94             | nd                | 164.42±16.58                   | nd                | 10.0      |
| 17  | 1.03±0.52             | 1.26±0.26         | 16.13±8.14                     | 19.74±4.07        | 7.5       |
| 22  | 3.60±0.66             | 0.63±0.14         | 47.75±8.75                     | 8.36±1.86         | 6.3       |
| 26  | 0.18±0.57             | 4.02±0.40         | 2.62±8.28                      | 58.43±5.81        | 7.5       |
| <u>May 30, GISP 2 to ATM, Bearing 204° True</u> |                       |                   |                                |                   |           |
| 1   | 7.86±0.69             | nd                | 93.68±8.22                     | nd                | 7.5       |
| 3   | 3.47±0.57             | 0.73±0.24         | 42.89±7.04                     | 9.02±2.97         | 6.5       |
| 8   | 5.34±0.46             | 0.15±0.20         | 29.08±2.51                     | 0.82±1.09         | 3.5       |
| 13  | 10.07±0.72            | 1.43±0.19         | 75.44±5.39                     | 10.71±1.42        | 3.0       |
| 18  | 3.16±0.67             | 2.38±0.32         | 22.58±4.79                     | 17.00±2.29        | 4.5       |
| 23  | 4.88±0.56             | 3.16±0.37         | 44.43±5.10                     | 28.77±3.37        | 6.0       |
| 28  | 4.28±0.60             | 0.64±0.18         | 15.23±2.13                     | 2.28±0.64         | 3.0       |
| <u>June 24, from ATM, Bearing 114° True</u>     |                       |                   |                                |                   |           |
| 0   | 34.45±1.46            | 2.17±0.29         | 40.74±1.73                     | 2.56±0.34         | 3.0       |
| 3   | 30.95±1.12            | 1.21±0.34         | 41.99±1.52                     | 1.64±0.46         | 2.5       |
| 6   | 39.68±1.34            | 1.91±0.25         | 37.40±1.26                     | 1.80±0.23         | 2.0       |

Here, nd means not detected.

\* Distances are measured from GISP 2 for May 30 and from ATM for June 24.

TABLE 3. Monthly Average Concentrations of <sup>7</sup>Be and <sup>210</sup>Pb During Midsummer at a Number of Polar Stations

| Site             | Period of Record | Latitude | Longitude | Altitude, m | <sup>7</sup> Be |             | <sup>210</sup> Pb |       | Reference |
|------------------|------------------|----------|-----------|-------------|-----------------|-------------|-------------------|-------|-----------|
|                  |                  |          |           |             | June            | July        | June              | July  |           |
| Dye 3            | 1989             | 65°10'N  | 44°44'W   | 2560        | 66              | 52          | 4.0               | 2.1   | 1         |
| Summit           | 1989             | 72°20'N  | 38°45'W   | 3220        | 123             | 82          | 7.2               | 4.0   | 1         |
| Nord             | 1971             | 81°40'N  | 17°00'W   | 250         | 44              | 42          |                   |       | 2         |
| Thule            | 1970-1975        | 76°36'N  | 68°35'W   | 259         | 57<br>(22)      | 58<br>(26)  |                   |       | 2         |
| Kap Tobin        | 1974-1980        | 70°25'N  | 21°59'W   | 22          | 30<br>(9)       | 26<br>(9)   |                   |       | 2         |
| Barrow           | 1976-1985        | 71°10'N  | 156°30'W  | 4           | 25              | 26          | 1-5*              | 1-5*  | 2,3       |
|                  |                  |          |           |             | Dec.            | Jan.        | Dec.              | Jan.  |           |
| South Pole       | 1970-1984        | 90°00'S  |           | 2800        | 173<br>(49)     | 179<br>(25) | 1-4+              | 1-4+  | 2,4       |
| GVN              | 1983-1987        | 70°37'S  | 8°22'W    | ?           | 100-200         | 100-200     | 1-1.5             | 1-1.5 | 5         |
| Dumont D'Urville | 1978-1980        | 66°40'S  | 140°01'E  | 40          | 207             | 200         |                   |       | 6         |

All activities in femtocuries per standard cubic meter. Standard deviation given in parentheses for stations with multiyear records (when known).

References: 1, this work; 2, EML data reports, as summarized by Feely et al. [1988]; 3, Rahn and McCaffrey [1980]; 4, Maenhaut et al. [1979]; 5, Wagenbach et al. [1988]; 6, Sanak et al. [1985].

\*<sup>210</sup>Pb data for 1983 - 1985 (reference 2) and 1976 -1978 (reference 3).

+<sup>210</sup>Pb data for 1976 -1977 (reference 4).

down to surface levels. (The similar <sup>7</sup>Be concentrations seen at the Antarctic stations may also be reflecting significant subsidence of upper atmospheric air masses within the polar vortex [Dibb et al., 1990; Lambert et al., 1990; Kamiyama et al., 1989; Wagenbach et al., 1988; Sanak et al., 1985].) The average <sup>210</sup>Pb concentrations over the Greenland Ice Sheet (Table 3) are near, or below, the low end of the range reported for continental sites (5-25 fCi m<sup>-3</sup> STP, compiled by Lambert et al., [1982]), reflecting the loss of submicron aerosols during long-range transport from the presumed major source areas of Eurasia and North America.

The rapid concentration fluctuations of <sup>7</sup>Be and <sup>210</sup>Pb in near-surface air over the Greenland Ice Sheet are intriguing, particularly given the close correspondence between these two radionuclides with distinctly different sources. Similar variability has been observed in Antarctica, where the high-concentration episodes were attributed to a combination of vertical mixing processes in the troposphere and stratospheric intrusions [Wagenbach et al., 1988; Sanak et al., 1985]. The frequent short-term occurrences of <sup>7</sup>Be levels at Summit approaching, or exceeding, 100 fCi m<sup>-3</sup> STP, superimposed on a background near 50 fCi m<sup>-3</sup> STP, are very similar to the detailed time series observed at White Face Mountain by Husain and coworkers during the 1970s. In that study, <sup>7</sup>Be spikes greater than 200 fCi m<sup>-3</sup> STP, against background

values near 100 fCi m<sup>-3</sup> STP, were clearly shown to result from stratospheric intrusions associated with tropopause folds [Dutkiewicz and Husain, 1979; Husain et al., 1977]. However, it has been suggested that at low to middle latitudes much of the variability in ground level <sup>7</sup>Be concentrations is due to changes in the intensity of vertical mixing in the troposphere [Feely et al., 1988; Graustein and Turekian, 1989]. For several coastal sites in the Arctic, Feely et al. [1988] suggest that much of the <sup>7</sup>Be is rapidly advected through the troposphere from mid-latitude regions with higher concentrations and that short intervals of elevated <sup>7</sup>Be reflect particularly effective transport episodes. The relatively high elevations of the sampling sites on the ice sheet (Figure 1, Table 3), and the lower altitude of the tropopause in polar regions (8-10 km [Reiter, 1975]), suggest that some, if not most, of the upward excursions in the <sup>7</sup>Be time series presented here may also be reflecting stratospheric air masses descending to the surface.

The common occurrence of peaks in ground level <sup>210</sup>Pb concentration coinciding with <sup>7</sup>Be peaks (Figure 2) might support the notion of a tropospheric source for the <sup>7</sup>Be peaks. However, it has been suggested that much of the <sup>210</sup>Pb reaching Antarctica is transported through the stratosphere [Lambert et al., 1966; Maenhaut et al., 1979; Wagenbach et al., 1988]. The frequent short episodes of elevated levels of

$^7\text{Be}$ ,  $^{210}\text{Pb}$ , and  $^7\text{Be}/^{210}\text{Pb}$  (Figures 2 and 4) in the air over the Greenland Ice Sheet may reflect stratospheric intrusions through tropopause folds associated with the polar jet stream [Raatz et al., 1985; Shapiro et al., 1987] or the passage of fronts [Wolff et al., 1979; Dutkiewicz and Husain, 1979; Husain et al., 1977]. Lidar measurements during the NASA ABL 3A mission over Alaska in the summer of 1988 frequently showed isolated stratospheric air masses below the tropopause [Browell et al., 1989]. These pockets of stratospheric air appeared to have "broken off" an irregular tropopause and were sometimes seen to reach down to the surface. A similar occurrence was observed during one of the three AGASP 2 flights over Alaska in early April 1986 [Herbert et al., 1989]. The paucity of upper atmospheric data over the Greenland Ice Sheet currently precludes assessment of the frequency or efficiency of any of these processes that could bring stratospheric material to the surface of the ice sheet. Careful analysis of the complete DGASP data set, including all available meteorologic information, together with data on the vertical distribution of  $^7\text{Be}$  and  $^{210}\text{Pb}$  over northeastern Greenland during March 1989 (from the AGASP 3 mission (J.E. Dibb, manuscript in preparation, 1990) should help resolve the relative importance of stratospheric and tropospheric sources at ground level over the Greenland Ice Sheet. If most of the  $^7\text{Be}$  is indeed stratospheric, it would appear that  $^{210}\text{Pb}$  also has a significant stratospheric component in its long-range transport. This would imply that many other continentally derived atmospheric constituents, including anthropogenic pollutants, may sometimes follow a stratospheric pathway to the ice sheet as well.

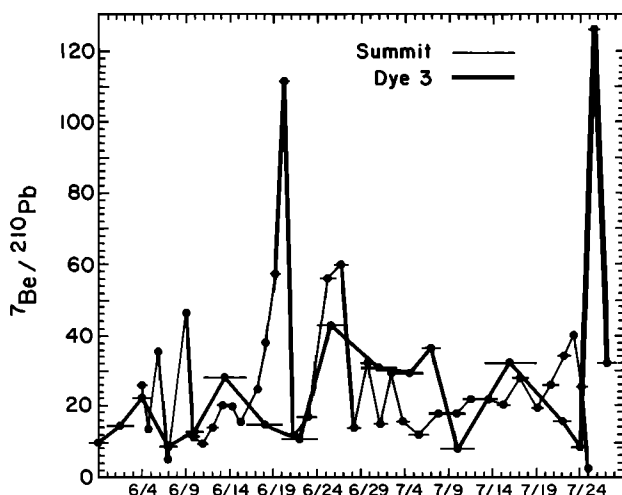


Fig. 4. Comparison of  $^7\text{Be}/^{210}\text{Pb}$  in surface level air at Summit and Dye 3 during June and July 1989.

#### Air-Snow Relationships

The comparable concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in fresh snowfall at Dye 3 and Summit, despite aerosol concentrations being nearly 2 times higher at Summit, is surprising. This might indicate that the processes removing submicron-aerosol-associated radionuclides from the atmosphere to the snow are more effective over Dye 3 than Summit and is certainly reflected in the higher scavenging ratios (calculated as (concentration in snow  $\times$  density of air) / concentration in air) at Dye 3 (Table 1). However, it is quite likely that scavenging ratios derived from ground level aerosol concentrations are a crude oversimplification of very complicated processes. Perhaps the biggest problem with

such ratios is the fact that the snowflakes are generally formed in air masses several kilometers above the surface. Data obtained from the AGASP 1 and 2 aircraft missions have shown that stable temperature inversions over the Arctic ice pack and snow-covered land in the winter limit downward mixing so effectively that the concentrations of many atmospheric constituents are greatly reduced in boundary layer air relative to the air just above the inversion [Oltmans et al., 1989; Mickle et al., 1989]. Although the presence and persistence of such conditions have not been examined as part of the present investigation, they would certainly limit the usefulness of scavenging ratios calculated from ground level atmospheric concentrations. Perhaps the snow falling at Summit and Dye 3 is formed in a homogeneous air mass above such an inversion, and the higher  $^7\text{Be}$  and  $^{210}\text{Pb}$  concentrations in ground level air at Summit reflect a thinning of the boundary layer due to topographic or orographic effects. However, the different temporal trends in ground level air concentrations at the two sites (Figures 2 and 3) indicate the situation is more complex than this, perhaps including patches of stratospheric air in the troposphere (as were seen over Alaska) above a surface inversion layer and/or short-term, localized breakdown of the inversion resulting in variable ventilation of the boundary layer.

Scavenging ratios calculated for the two rime/frost samples collected at Summit yield values for  $^7\text{Be}$  of 860 and 700, while the  $^{210}\text{Pb}$  ratios were 2150 and 1240 for the July 4 and 5 samples, respectively. The material collected on the vertical screen is apparently derived from ground level air, so these scavenging ratios may be more realistic. Direct comparison to the ratio values for snow (Table 1) indicate that the rime may be more effective than snow at removing submicron aerosols from the air over the Greenland Ice Sheet. It should be noted that the postulated presence of a depleted boundary layer under a stable near-surface inversion would make the scavenging ratio values in Table 1 erroneously high, and a true comparison between values for snow and rime would show an even larger difference. It is unclear how much of this rime would be deposited in the absence of a large vertical obstruction, so the potential importance of such material to the record preserved in the snowpack is uncertain. Davidson et al. [1987, 1989] have suggested that the incorporation of rime into growing snow crystals during summer months contributes substantially to the annual deposition of sulfate at Dye 3; perhaps this mechanism is also important for the atmospheric radionuclides. However, examination of snowflake replicates collected throughout DGASP indicate little or no rime of the snow at Dye 3 at any time of the year (R. Borys, personal communication).

#### Atmospheric Fluxes of $^7\text{Be}$ and $^{210}\text{Pb}$

An estimate of the wet deposition fluxes of  $^7\text{Be}$  and  $^{210}\text{Pb}$  to the surface at Summit during June and July was made by summing the input from each of the fresh snow events (Table 1). If conservative estimates of 30 pCi  $^7\text{Be}$   $\text{m}^{-2}$  and 3 pCi  $^{210}\text{Pb}$   $\text{m}^{-2}$  are made for the July 10 sample, the total accumulation was 300 pCi  $^7\text{Be}$   $\text{m}^{-2}$  and 35 pCi  $^{210}\text{Pb}$   $\text{m}^{-2}$  for the 53-day period. These estimates correspond to average daily fluxes of 5.7 and 0.7 pCi  $\text{m}^{-2}$  for  $^7\text{Be}$  and  $^{210}\text{Pb}$ , respectively. Recent results from four snowpits on the Greenland Ice Sheet [Dibb, 1990] and a 50-m core recovered at Summit in 1989 (J.E. Dibb, "The accumulation of  $^{210}\text{Pb}$  at Summit, Greenland since 1855," submitted to *Tellus*, 1990; hereafter cited as Dibb, submitted) indicate that there is no discernible seasonality in the accumulation of  $^{210}\text{Pb}$  on the Greenland Ice Sheet. The lack of seasonality in  $^{210}\text{Pb}$  accumulation suggests that the wet deposition fluxes of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in June and July may be representative of a constant flux throughout the year, allowing estimation of

annual fluxes of  $^7\text{Be}$  and  $^{210}\text{Pb}$  of 1800 and 210 pCi  $\text{m}^{-2}$ , respectively. Alternatively, the 43 kg  $\text{m}^{-2}$  of snow deposited at ATM during June and July is 19.5% of the 74 year average annual accumulation of snow at the GISP 2 site (Dibb, submitted). Assuming a direct dependence of the wet deposition fluxes of  $^7\text{Be}$  and  $^{210}\text{Pb}$  on the amount of snowfall yields estimated annual fluxes of 1550 and 180 pCi  $\text{m}^{-2}$ , respectively. Both of these approaches to estimating annual flux probably err on the low side, as the aerosol concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  at Dye 3 during June and July were among the lowest observed during the entire DGASP campaign.

Estimation of wet deposition at Dye 3 is highly uncertain due to the limited information recorded for the fresh snow samples. The values in Table 1 are based on the range in depth and density of each snowfall event, determined at an accumulation stake 1 km east of the station. The total wet deposition during June and July was probably in the range of 375-740 pCi  $^7\text{Be}$   $\text{m}^{-2}$  and 27-53 pCi  $^{210}\text{Pb}$   $\text{m}^{-2}$ . These values yield estimated daily fluxes of 7-14 and 0.5-1.0 pCi  $\text{m}^{-2}$  of  $^7\text{Be}$  and  $^{210}\text{Pb}$ , respectively. The apparently much higher flux of  $^7\text{Be}$  due to wet deposition at Dye 3 compared to Summit may be important, but the uncertainty of the estimate calls the validity of this observation into question.

Davidson et al. [1987] suggest that monitoring temporal changes in the concentration of chemical species in fresh snow as it ages would provide an estimate of their dry deposition flux. Increasing concentrations in snow might also reflect simple mass losses due to sublimation of the snow, so that such estimates must be regarded as maximum values if sublimation is not considered. An attempt to estimate the dry deposition of  $^7\text{Be}$  and  $^{210}\text{Pb}$  at Summit was made by sampling snow on June 18 that fell June 13 (see Table 1). The inventories of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in the surface layer of snow increased to 37.7 and 10.8 pCi  $\text{m}^{-2}$ , respectively, suggesting daily dry deposition fluxes of 4.8 pCi  $^7\text{Be}$   $\text{m}^{-2}$  and 1.5 pCi  $^{210}\text{Pb}$   $\text{m}^{-2}$  during this time interval.

These results suggest that dry deposition is comparable to wet deposition in delivering submicron aerosols to the Summit region of the Greenland Ice Sheet, at least during the summer. However, several lines of evidence indicate that such an interpretation is premature. The same approach to estimating dry deposition has been applied to 20 sampling pairs during DGASP with very inconsistent results; often the concentration of one radionuclide increased while the other decreased, or they both decreased. It appears that sampling difficulties and the inherent small-scale variability of surface snow chemistry severely limit the applicability of this approach. In addition, simple regression of annual  $^{210}\text{Pb}$  accumulation against annual snow accumulation for the 134-year record recovered from the GISP 2 site suggests no significant dry fall component for  $^{210}\text{Pb}$  (Dibb, submitted). Finally, the average annual  $^{210}\text{Pb}$  accumulation at the GISP 2 site has been 165 pCi  $\text{m}^{-2}$  for the past 74 years and only 147 pCi  $\text{m}^{-2}$  since 1959. The annual  $^{210}\text{Pb}$  flux estimated from wet deposition alone during June and July 1989 (180 or 210 pCi  $\text{m}^{-2}$ , which are probably low estimates) exceeds the total annual accumulation of  $^{210}\text{Pb}$  observed for 30 of the last 35 years (Dibb, submitted).

Regardless of the true magnitude of the dryfall component, it is apparent that the depositional fluxes of  $^7\text{Be}$  and  $^{210}\text{Pb}$  are very low along the crest of the Greenland Ice Sheet. If, for illustrative purposes, a dry fall component of 50% is assumed, the annual fluxes of  $^7\text{Be}$  and  $^{210}\text{Pb}$  would be estimated at 3100-3600 and 360-420 pCi  $\text{m}^{-2}$ , respectively, at Summit and 4500-8880 and 320-640 pCi  $\text{m}^{-2}$  at Dye 3. Measured fluxes at mid-latitude northern hemisphere continental sites range from 42,000 to 102,000 pCi  $^7\text{Be}$   $\text{m}^{-2}$   $\text{a}^{-1}$  and 4100 to 4700 pCi  $^{210}\text{Pb}$   $\text{m}^{-2}$   $\text{a}^{-1}$  [Olsen et al., 1985;

Turekian et al., 1983; Bleichrodt and Van Abkoude, 1963]. These low fluxes onto the ice sheet presumably reflect the low precipitation rates, coupled with the long transport paths from source areas in the case of  $^{210}\text{Pb}$ . However, it should be noted that Monaghan and Holdsworth [1990] report an average annual accumulation of  $^{210}\text{Pb}$  in an ice core from Mount Logan (60°34'N, 140°30'W, 5951 m) of 390 pCi  $\text{m}^{-2}$ , which falls right in the range of estimates for the Greenland Ice Sheet. In this case the low  $^{210}\text{Pb}$  flux is attributed to the fact that snow accumulating on the high-elevation glacier is essentially sampling aerosols from upper levels of the atmosphere [Monaghan and Holdsworth, 1990].

### Summary And Concluding Remarks

The correlation between  $^7\text{Be}$  and  $^{210}\text{Pb}$  concentrations in ground level air over the Greenland Ice Sheet suggests that these radionuclides will not provide a straightforward indication of the relative importance of stratospheric and tropospheric sources of the air over the ice sheet. It seems likely that most of the short term variability seen at ground level is due to interactions on daily or shorter time scales between injections of air masses from above, mixing and removal processes in the boundary layer. However, over seasonal, or perhaps monthly, time periods the concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in the air at Summit should help to resolve the major pathways bringing continentally derived material to the ice sheet.

The marked difference in aerosol concentrations between Summit and Dye 3, in conjunction with very similar snow concentrations, indicates previously unexpected complexity, both spatially and temporally, of atmospheric structure over the ice sheet. Apparently, the boundary layer over the ice is highly dynamic, demanding careful assessment of the results of any surface-based atmospheric sampling campaign. Current plans for the three remaining GISP 2 drilling seasons tentatively include operation of a tether sonde system, an echo sounder, and release of rawinsondes. Such a program, perhaps supplemented by some aircraft sampling, would greatly strengthen interpretation of data collected during the GISP 2 and DGASP atmospheric sampling campaigns and might facilitate interpretation of the records recovered from the GISP 2 and GRIP deep cores.

Fresh snow sampling in the Summit region of Greenland has clearly indicated the potential for wind to greatly modify the atmospheric signal captured in the snow over relatively small spatial scales. Comparison of the long-term record of  $^{210}\text{Pb}$  accumulation at the GISP 2 site with the observed deposition of  $^{210}\text{Pb}$  over a 2-month period suggests that the accumulation over yearly intervals is less than the sum of the input events delivering  $^{210}\text{Pb}$  to this site. Erosion of the surface snow by wind could easily create such a discrepancy, but this does cast doubt on the ability of snow and firn to preserve a high-resolution record of atmospheric inputs to the Greenland Ice Sheet. It is only through further detailed studies like DGASP and the atmospheric sampling component of GISP 2 that an accurate assessment of the degree to which glacial firn and ice records faithfully preserve various atmospheric signals will be possible.

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